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A STUDY OF THE MAGNETIC AND METALLURGICAL
PROPERTIES OF $Sm(Co,Fe,Cu,Zr)_z$
ALLOYS NEAR z = 8.5

FINAL REPORT

A.E. RAY

FEBRUARY 1984

PREPARED FOR:

U.S. ARMY RESEARCH OFFICE

PREPARED BY:

UNIVERSITY OF DAYTON
DAYTON, OHIO 45469

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20. ABSTRACT (Continue on reverse side if necessary and identify by block number)
Alloys corresponding to the compositions $Sm(Co_{0.50}Fe_{v}Cu_{0.04}Zr_{0.02})_{8.35}$ with $v = 0.23$ to 0.30 have been characterized magnetically and metallurgically. Very large intrinsic coercivities ranging from 28-32 kOe combined with saturation inductions of 11.6 - 12.1 kG have been obtained for the bulk alloys. If these properties can be incorporated into sintered permanent magnets, energy products of 26 - 33 MGOe should be possible for temperature compensated alloys, and 37 - 39 MGOe for uncompensated alloys.

(Block No. 20 continued)

The $Sm(Co_{0.690-x}Fe_xCu_{0.08}Zr_{0.02})_{8.35}$ alloys were found to behave metallurgically like alloys with similar transition metal content, but with z = 7.0 - 7.6. They form a single phase solid solution at high temperatures which is retained at room temperature by rapid quenching; on reheating to approximately 800°C, the high temperature phase dissociates by a spinodal-like reaction into a cellular microstructure consisting of 2:17 R cells, 1:5 cells and a platelet phase. All phase boundaries are coherent. The platelet phase was determined to have the "2:17 H" crystal structure but also Sm-depleted of 2:17 stoichiometry. By comparing lattice constants of the multicomponent alloys with those of $Sm_2(Co_{1-x}Fe_x)_{17}$ alloys at the same Fe levels, it was determined that Zr-vacancy pairs substitute for Fe pairs at the dumbbell sites in the 2:17 R phase at high temperatures.

A model for the metallurgical behavior of $Sm(Co,Fe,Cu,Zr)_z$ alloys between z = 7.0 - 8.5 has been proposed.

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PROBLEM STATEMENT

Magnetic circuits under consideration for micro- and milli-
meter wave tubes call for permanent magnets with energy products
greater than 30 MGOe combined with nearly zero reversible
temperature coefficients.[1] Such magnets are not presently
available. Nearly zero temperature coefficients can be achieved
in 2:17 type $Sm(Co,Fe,Cu,Zr)_z$ permanent magnets by the partial
substitution of Er for Sm, but at a sacrifice of 15 - 30% of
the available energy product.[2] The potential energy products
of uncompensated 2:17 type permanent magnets are in the 44 -
48 MGOe range, which should be sufficient to permit temperature
compensation by heavy rare earth substitution. However, the
development of 2:17 type permanent magnets with energy products
above 30 - 33 MGOe has so far proved an illusive goal. The
highest energy product reported for a 2:17 type permanent magnet
is 33 MGOe for $Sm(Co_{.65}Fe_{.28}Cu_{.05}Zr_{.02})_{7.7}$ by TDK Electronics
in 1980.[3] They also reported that all attempts to raise
remanent magnetization by increasing Fe or decreasing Cu, Zr,
or Sm contents resulted in serious loss of coercivity.

Shimoda, et al.[4] prepared bonded magnets made from alloys
corresponding to the composition $Sm(Co_{.90-v}Fe_vCu_{.08}Zr_{.02})_z$
with $v = 0.22 - 0.30$ and $z = 8.35$. In contrast to the results of
TDK, they obtained coercivities up to 18 kOe in their bonded
magnets. They did not report on the bulk metallurgical or
magnetic properties of their nearly stoichiometric alloys, but
with saturation magnetizations potentially 6% to 9% higher
than the TDK alloys with $z = 7.4 - 7.6$, energy products of
$(BH)_{max} = 37 - 39$ MGOe might be possible with such alloys.

The primary emphasis of this program was to study the
basic metallurgical and magnetic properties of the
$Sm(Co_{.90-v}Fe_vCu_{.08}Zr_{.02})_{8.35}$ alloys and to evaluate their
potential for high energy sintered permanent magnets. We also
planned to study the coercivity mechanisms of these alloys,
which were expected to differ significantly from the alloys

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with z = 7.4 - 7.6. Finally, we hoped to develop a better understanding of the metallurgical behavior of these complex alloy systems in order to develop improved high energy permanent magnet alloys.

SUMMARY OF IMPORTANT RESULTS

The hard magnetic properties and some metallurgical properties are presented and discussed in [5]. The most important magnetic results are summarized in Figures 1 and 2 and in Table I. Saturation magnetization, $4\pi M_s$, values obtained in this study are compared with those of Shimoda in Figure 1. Intrinsic coercivities, $_MH_c$, are compared in Figure 2. The values of $_MH_c$ we obtained by various aging treatments are given in Table I. These results have lead us to conclude that

(1) Very high values of $4\pi M_s$ and $_MH_c$, much higher than those reported by Shimoda, can be developed in these alloys.

(2) The large drop in $_MH_c$ for similar alloys with z>7.6 does not apply to alloys with z = 8.35.

(3) If the magnetic properties of the bulk alloys can be incorporated into sintered permanent magnets, energy products in the range of 37 - 39 MGOe should be possible with these uncompensated alloys. With Er partially substituting for Sm[2] temperature compensated permanent magnets in the range of 26 - 33 MGOe may be attainable.

Two very important results pertaining to the metallurgical behavior of the $Sm(Co_{.90-v}Fe_vCu_{.08}Zr_{.02})_{8.35}$ alloys were obtained in this study. These are shown in Figures 3 and 4. Figure 3 shows the TEM microstructure of a solutionized and aged alloy. It clearly has the cellular microstructure observed for similar alloys with z = 7.0 - 7.6. From this and similar micrographs we can conclude:

(1) The coercivity mechanisms for the z = 8.35 alloys must be very similar to those for the z = 7.0 = 7.6 alloys.

(2) The thickness of the 1:5 cell boundary phase, relative to the size of the 2:17 R cells, is largely determined by the value of z.

(3) The platelet phase must have the 2:17 H crystal structure, and must be quite Sm-poor of the 2:17 stoichiometry. Figure 4 compares the lattice parameters of the $z = 8.35$ alloys in the solid solution heat treated (SSHT) and aged conditions with those of ternary $Sm_2(Co_{1-x}Fe_x)_{17}$ at the same iron contents. From these measurements we have concluded that:

(1) Zr, as Zr-vacancy pairs, displace Fe pairs from the dumbbell positions in the 2:17 R phase at high temperatures.

(2) The function of Zr is to stabilize the 2:17 R phase by preventing Co-Fe phase precipitation during isothermal aging.

During the final stages of the work effort we began to develop a model for the metallurgical behavior of the multi-component $Sm(Co,Fe,Cu,Zr)_z$ alloys. These alloys are much too complex for cut and try approaches to have much success in improving their magnetic properties. The model in its early stage of development is described in [6] and as it presently stands in [7]. Our future efforts to develop permanent magnets which combine excellent temperature compensation with energy products well above 30 MGOe will revolve around the testing and refinement of our model for the metallurgical behavior of $Sm(Co,Fe,Cu,Zr)_z$ alloys.

Figure 1.

Comparison of $4\pi Ms$ values for
$Sm(Co_{.90-v}Fe_vCu_{.08}Zr_{.02})_{8.35}$ alloys
isothermally aged 24 hrs. at 800° C.

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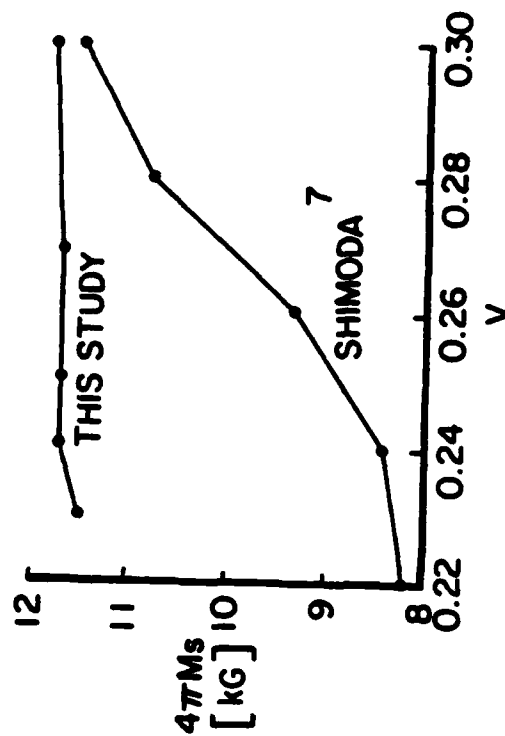
Figure 2.

Comparison of $_MH_C$ values for
Sm (Co$_{.90-v}$Fe$_v$Cu$_{.08}$ Zr$_{.02}$)$_{8.35}$ alloys
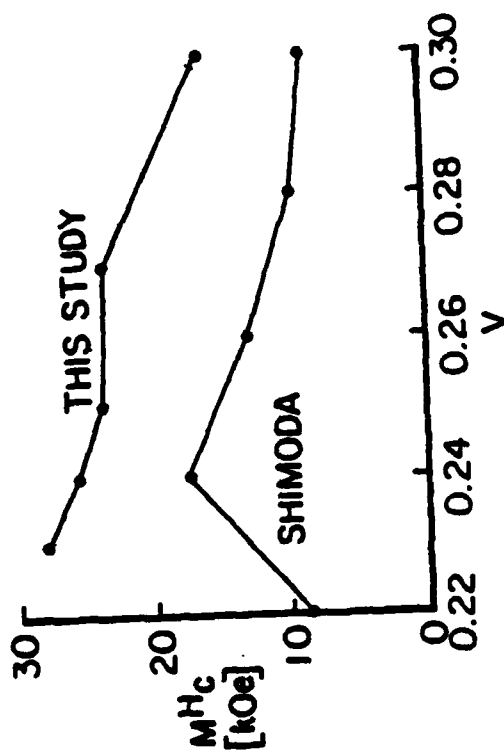isothermally aged 24 hrs. at 800°C.

TABLE I

Comparison of Rapid Cooling (Q) and Furnace Cooling (FC) after Isothermal Aging at 800°C on Intrinsic Coercivity, $_MH_C$, of Sm (Co$_{.90-v}$ Fe$_v$ Cu$_{.08}$ Zr$_{.02}$) 8.35 Alloys

AMT. Fe v	SSHT °C	8h+Q $_MH_C$, kOe	8h+FC* $_MH_C$, kOe	24h+Q $_MH_C$, kOe	24h+FC* $_MH_C$, kOe
0.23	1170	3.5	6.2	28.0	20.0
0.23	1160	3.6	5.1	24.4	21.8
0.24	1170	11.0	10.3	25.5	29.5
0.24	1160	11.2	13.8	25.2	22.0
0.25	1160	16.1	17.7	24.2	30.5
0.25	1150	16.4	27.0	23.8	30.2
0.27	1160	8.4	9.4	23.5	31.3
0.27	1150	15.5	18.5	23.6	31.6
0.30	1130	5.5	6.7	16.2	19.8
0.30	1120	10.1	11.4	14.4	21.9

FC* = 2°C/min. from 800°C to 400°C

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Figure 3.

TEM Microstructure of Sm(Co$_{.65}$Fe$_{.25}$Cu$_{.08}$Zr$_{.02}$)$_{8.35}$ Alloy
Solid Solution Heat Treated at 1150C for 24h, then AGED at 800C for 8h.
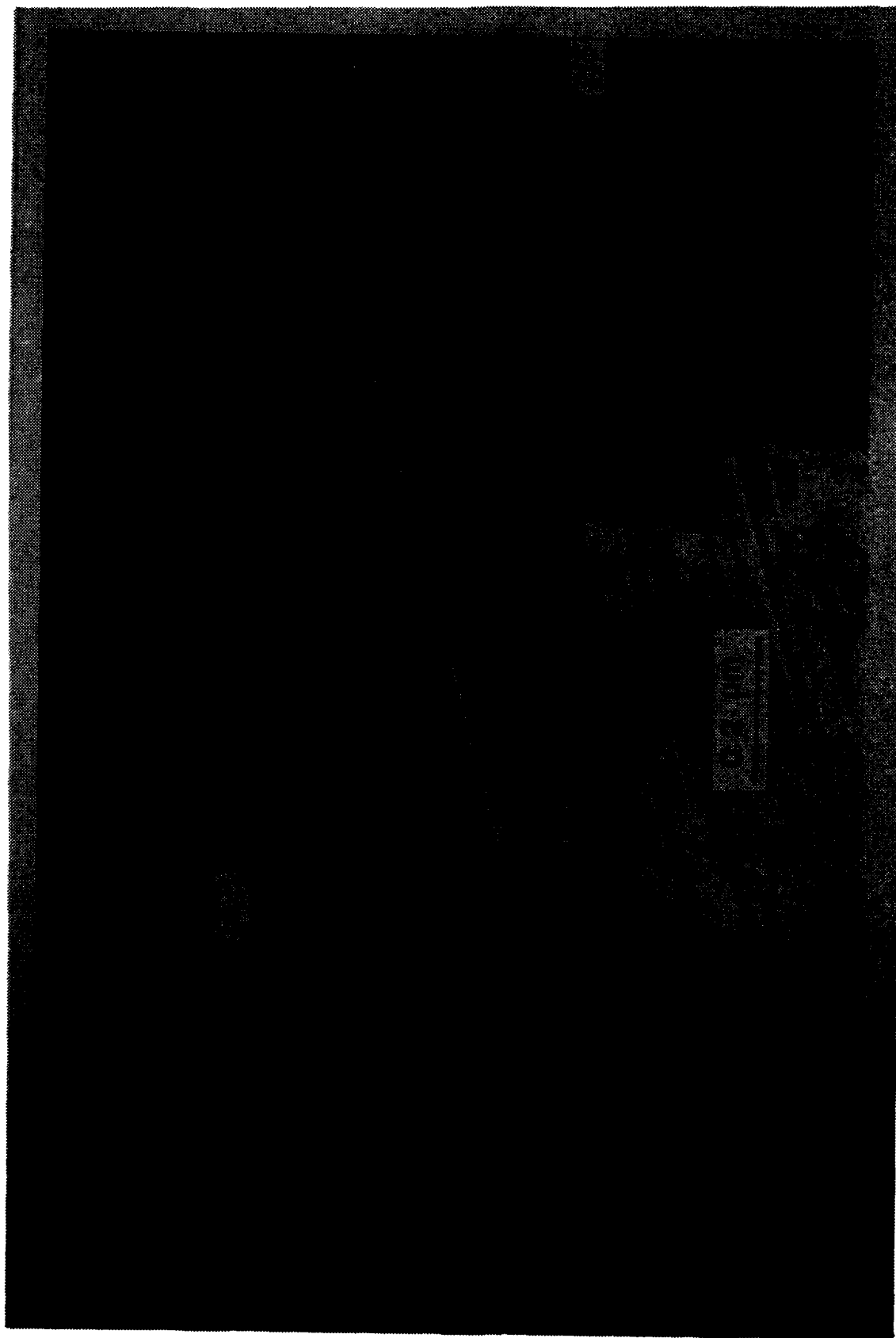
TEM Micrograph by George Hadjipanayis

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Figure 4. Lattice Constants

$Sm(Co_{.90-v}Fe_vCu_{.08}Zr_{.02})_{8.35}$
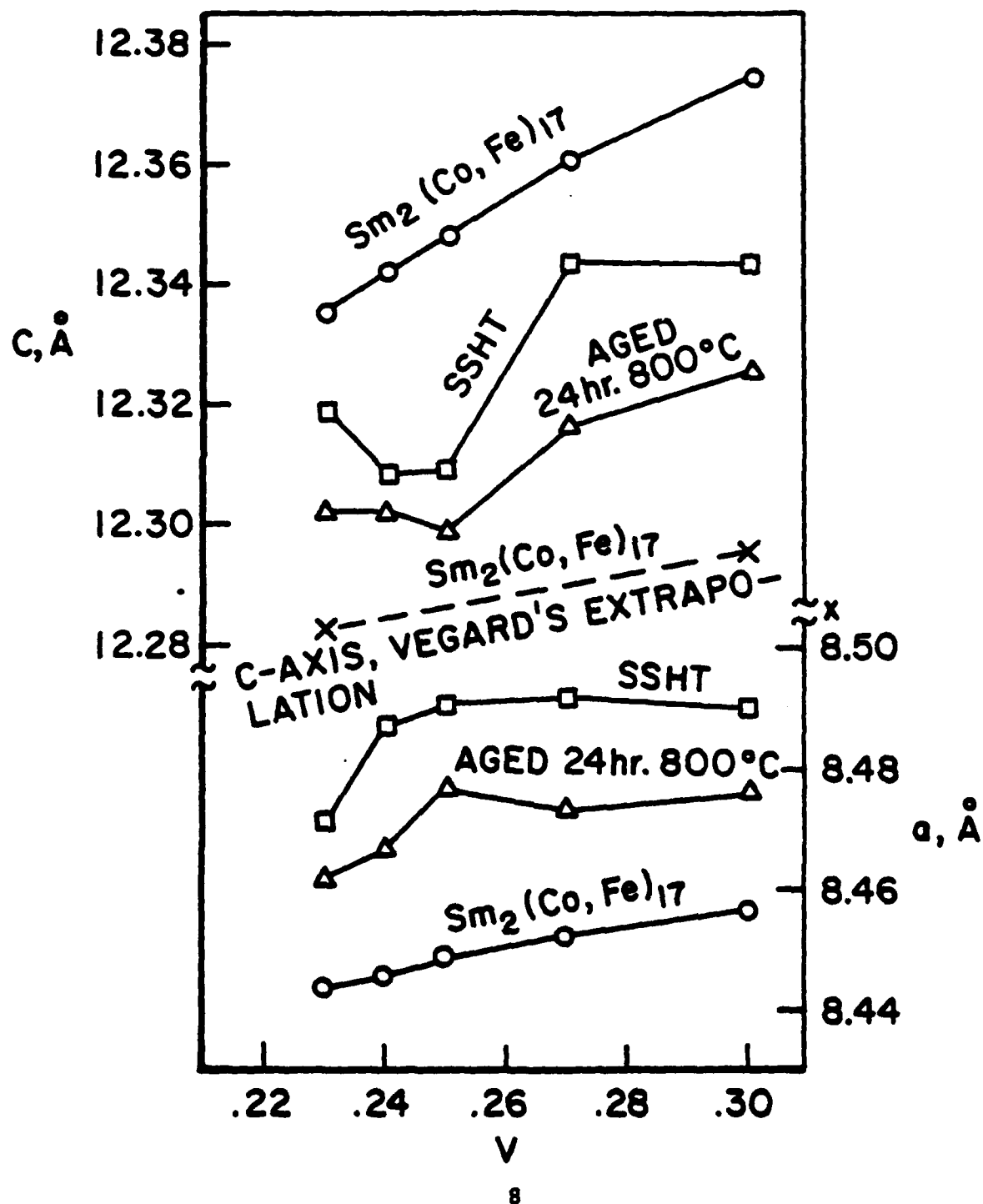
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1. A. E. Ray, H. F. Mildrum, and J. B. Krupar, "Magnetic and Metallurgical Properties of $Sm(Co,Fe,Cu,Zr)_z$ Alloys Near $z = 8.5$," IEEE Trans. Magnetics, MAG-19 (1983) 2044-46.

2. H. F. Mildrum, J. B. Krupark, and A. E. Ray, "High Coercive Force 2-17 Type $Sm_{1-x}Er_x(Co_{.69}Fe_{.22}Cu_{.08}Zr_{.02})_{7.22}$ Magnets With a Low Temperature Coefficient," J. Less-Common Metals, 93 (1983) 261-267.

3. A. E. Ray, "A Model for the Metallurgical Behavior of $Sm(Co,Fe,Cu,Zr)_z$ Alloys in the Development of Hard Magnetic Properties," Seventh International Workshop on Rare Earth-Cobalt Permanent Magnets, Beijing, China, September, 1983 (China Academic Publishers) pp. 261-268.

4. A. E. Ray, "The Metallurgical Behavior of $Sm(Co,Fe,Cu,Zr)_z$ Alloys." Paper EB-04 presented at 29th MMM Conference, Pittsburgh, PA, 8-11 November 1983. Accepted for publication in J. Appl. Physics, Spring, 1984.

5. K. J. Strnat, D. Li, and H. F. Mildrum, "Magnetic Domains and Reversal Mechanisms in Sintered "$SmCo_5$" Permanent Magnets." Paper EB-06, Ibid.

6. D. Li and K. J. Strnat, "Domain Structures of Two Sm-Co-Cu-Fe-Zr "2-17" Magnets During Magnetization Reversal." Paper EB-07, Ibid.

PARTICIPATING SCIENTIFIC PERSONNEL

The following scientific personnel participated in the research effort:

1. Alden E. Ray, (Principal Investigator) Professor of Materials Engineering and Supervisor of Metals and Ceramics Division of the Research Institute.

2. Karl J. Strnat, Tait Professor of Electrical Engineering and Director of the Magnetics Laboratory.

3. Herbert F. Mildrum, Adjunct Professor of Electrical Engineering and Research Engineer in the Research Institute.

4. Richard S. Harmer, Assoc. Professor of Materials Engineering, Research Engineer, and Head of the Materials Analysis Laboratory, Research Institute.

No advanced degrees were earned by anyone particpating in this project.

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6. A. E. Ray, Proc. 7th International Workshop on REPM and Their Applications, Beijing, China, 261 (1983).

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